

Research interest: Theory of Nanomaterials

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I present a summary of my research experience on nano-science, in particular, on semiconductor nanostructures. I give a perspective on the current state of the field and my personal views on promising areas for research and discuss some my plans for the future.

INTRODUCTION

Since I started my dissertation work at Instituto Balseiro Argentina I was captivated by the possibility of controlling artificial materials at the nanoscale. At that time the field was only emerging; far from national initiatives and specialized centers. Thirteen years after, nano-science is a multidisciplinary research subject going far beyond condensed matter to involve chemists and biologists in every corner of the world. This is a field where flexibility matters, a combination of two or more techniques is often required, and the ability to interact to with a diverse team is increasingly important. There is a realm of new experiments coming every day. Those experiment drive us, theoreticians, to stretch, to go a little bit further and to learn something new every day. I have to say that it is fun. My perspective is that the field will continue to grow and there is room for all. So if you are reading this because you are looking for a job and want to know what we do, please go ahead and send us your resume. I promise, if you work with us you are not going to be bored. Coming back to my research interest I am interested both in theoretical methods and the applications of those methods to solid state.

Along my research path I have gathered experience in several of the theoretical techniques applied to electronic structure. Of course I have not covered all. The applications have been concentrated in the confined electron gas, semiconductor nanostructures (quantum wells, wires and dots) and semiconductor surfaces. Currently I am exploring some ideas in nanotubes and metals.

GRAPHICAL CURRICULUM VITAE

In the rest of this document I present a collection of figures from papers of the past and some of publications to be. They constitute a sort of graphical C.V. The rest of the text is a summary of my research accomplishments and plans for the immediate future. It is oriented to the ones that wish more information than the one provided in my CV and less than the one provided in my publications. **For a concise description, please refer to the first two pages of my C.V.**

My dissertation work was under the advice of C. R. Proetto. The subject of that work was semiconductor super-lattices.

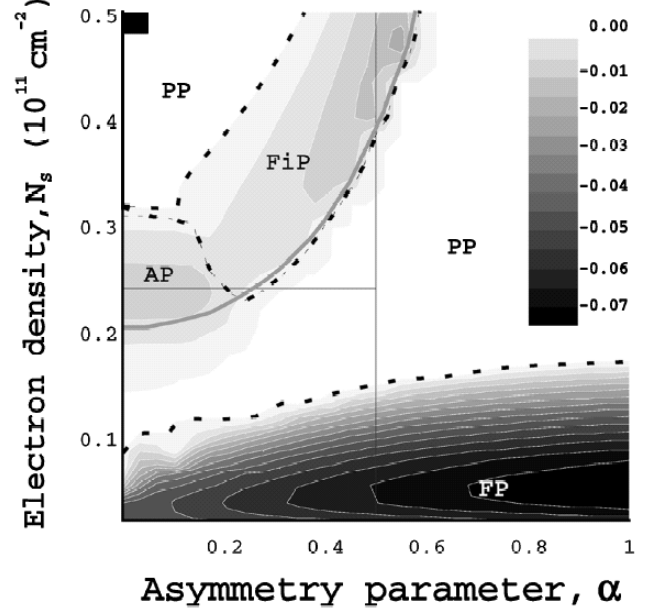


FIG. 1: Magnetic phase diagram of a quasi-two-dimensional-electron gas in a double quantum well as a function of the total density and asymmetry of the system. Self consistent DFT calculation as reported in Ref. [3].

The central problem in that work was the self consistent treatment of electron[1] and hole[2] gases which we carried out within the effective mass (EMA) and $\mathbf{k}\cdot\mathbf{p}$ approximation using density functional theory (DFT). We proposed the first rough approximation for the exchange correlation of holes[2] that initiated the theoretical debate in this area.

In 1992 I started my Ph.D. thesis, also with César Proetto. I studied the electron gas in semiconductor quantum wires[4–6] and wells[3, 7] including ground state and optical properties with DFT and time dependent DFT (TDDFT) approaches (See Fig. 1). In addition, we studied nano-rings within a semi-classical hydrodynamical approach[8, 9]. I also concluded and published independently a classical Monte Carlo project on the spatial order of excited states in superlattices[10].

While my experience in DFT had not included ab-initio atomistic calculations, it helped me to obtain a Research Associate position with Sokrates Pantelides at Vanderbilt University. In 1997 I move to Nashville to work with him. We published two articles one on the

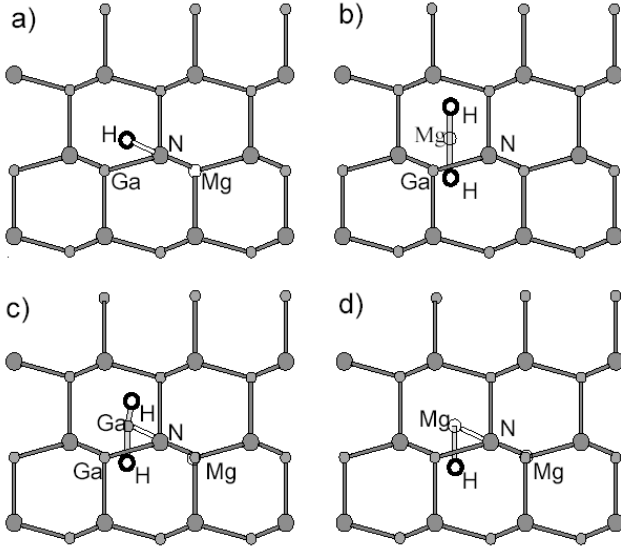


FIG. 2: Amphoteric chemistry of Mg dissolved in GaN. Several complexes of Mg interstitial (donor) and Mg substitutional (acceptor) pairs are found to be stable. When combined with H, they produce Raman modes that correlate well with unexplained experiments (See Ref. [11] for details).

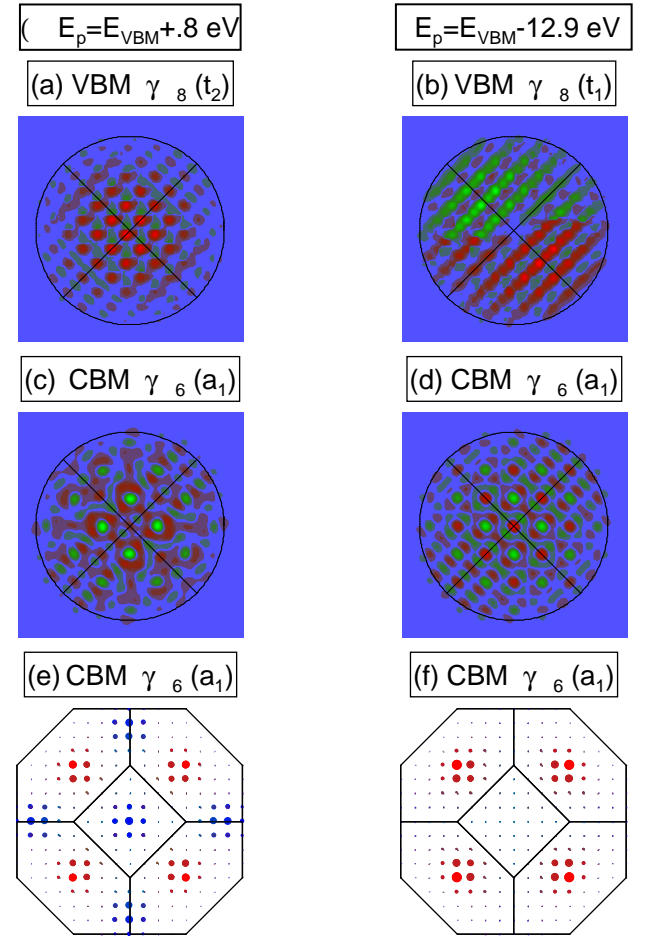
formation of H platelets[12], and the other on the effects of H in Mg doping of GaN[11] (See Fig. 2).

In 1998 I moved to Colorado to work with Alex Zunger, I wanted to learn the empirical pseudopotential method (EPM) developed in his group, in particular, the work involving Lin-Wang Wang (now at LBNL). This method covers the size range between EMA or $\mathbf{k}\cdot\mathbf{p}$ methods and ab-initio calculations. I worked in group IV semiconductor quantum dots publishing a number of papers. Four of them were EPM calculations and configuration interaction (CI) calculations of Si[13, 14] and Ge[15, 16] quantum dots. One of these calculations revealed the importance of the surface passivation on quantum dots even in the large size regime[16] (See Fig. 3). This stimulated an ab-initio study of the effects of H passivation on Si surfaces and steps in particular[17].

In 2000 I returned to Argentina with a permanent research position from CONICET (National Council for research in Science and Technology). Far from large computers I choose to do mostly analytical theory in quasi two dimensional electron gases (2DEG). With the perspective of ab-initio calculations in mind we did research in energy functionals which are at the core of every ab-initio DFT code. We successfully derived an exact exchange theory for the 2DEG without approximations[18, 19] (See Fig. 4). With our theory we could explain[20] first order phase transitions observed experimentally[20] and predict others[21]. Our prediction of spin polarized phases was recently verified experimentally by Alejandro Goñi's group in Berlin Germany[21]. I was also involved in TDDFT calculations on the collective modes of spin

polarized quantum wells[22].

In 2001 I decided to emigrate permanently to the USA and Since then I am working as a term staff in the quantum simulation group at Lawrence Livermore National Laboratory led by Giulia Galli. Over the years much of my research has pointed me to consider the properties of the surfaces. Technical reasons limit these calculations to the largest supercomputers and the best codes. Therefore, here in LLNL I concentrated on the chemistry of quantum dots surfaces, which is particularly important for sensors and biological applications. I am contributing to a LLNL effort to describe with ab-initio methods the effects of the surface structure and chemistry on quantum dots (See Fig. 5). The group has produced a number of high profile articles on the subject. I have authored some of them[23–25].



Reboredo and Zunger COLOR Figure 1

FIG. 3: Drastic changes on the shape and symmetry of the band edge wavefunctions of a Ge quantum dot as a function of the surface passivation. This EPM calculations (See Ref. [16]) stimulated detailed ab-initio calculations investigating the influence of the surface on quantum dots on their optical properties.

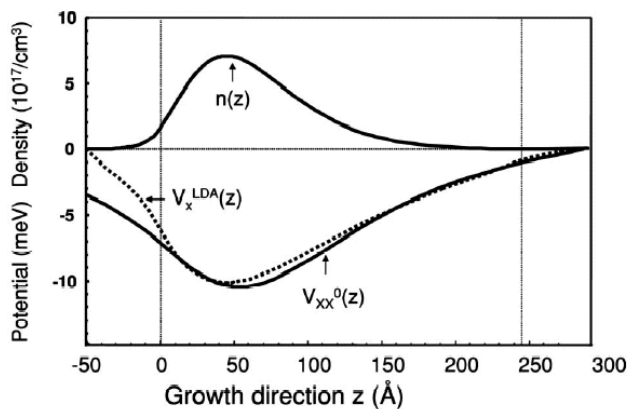


FIG. 4: Comparison of the exact exchange potential with the LDA potential for a quasi two dimensional electron gas when only one subband is occupied. The full upper line is the 3D density $n(z)$ obtained with XX theory[18] corresponding to a 2D density of $0.683 \cdot 10^{12}/\text{cm}^2$. XX and LDA exchange potentials are compared for this same density. Energies are measured from the chemical potential μ .

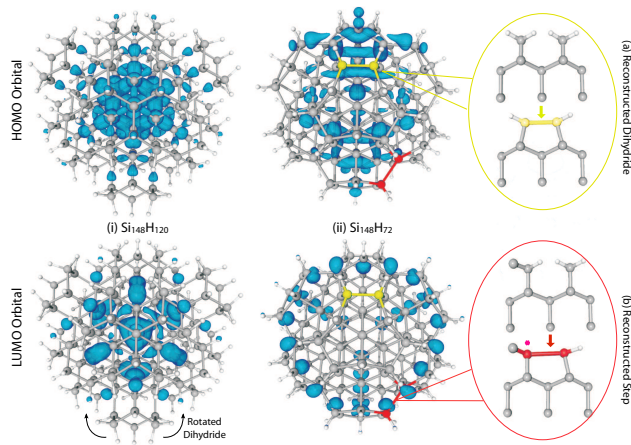


FIG. 5: Ab-initio calculation of the surface reconstruction effects on Si quantum dots. Unreconstructed ($\text{Si}_{148}\text{H}_{120}$) and reconstructed ($\text{Si}_{148}\text{H}_{72}$) hydrogen (white atoms) passivated 1.8 nm silicon (grey atoms) nanoclusters. Charge density iso-surfaces (blue) represent 50

In addition, I have done calculations explaining optochemical reactions in porous Silicon[26](See Fig. 6).

Currently I am studying the effects of a full organic termination on the optical and structural properties of Si and Ge quantum dots (Fig. 7).

I have also learned quite a bit from the group. Some of the codes we use, developed by Francois Gygi, are specialized to perform large Car-Parinello simulations (which is a unified ab-initio molecular dynamics approach). Currently, we are taking advantage of these codes to design structures for hydrogen storage[25].

During my stay here I could not avoid taking advantage of the presence of a number of experts on quantum Monte Carlo (QMC) methods and I am currently

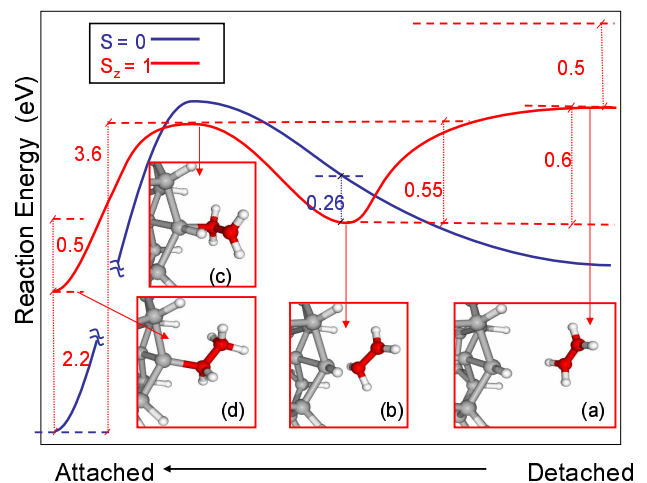


FIG. 6: Optochemistry of Si dots: Schematic representation of the reaction $\text{Si}_{29}\text{H}_{24} + \text{C}_2\text{H}_4 \rightarrow \text{Si}_{29}\text{H}_{23}\text{-CH}_2\text{-CH}_3$ in the excited state ($S_z = 1$, continuous red line). The energy values for the ground (singlet) configuration are also shown for comparison ($S = 0$, blue line). Note that the energy barriers are drastically reduced in the excited state. The atomic configurations of the reactants, product and transition states are shown in the insets. See Ref. [26] for details.

working on the method and learning it the same time. Until now QMC was restricted to a small group of physicists working with few atoms. But Andrew Williamson, Randy Hood and Jeffrey Grossman recently developed a linear scaling algorithm for QMC calculations based on a unitary transformation. [28]. This improvement, which allows the calculation of optical gaps and total energies of quantum dots as large as 250 atoms, obviously attracted my curiosity (It is predictable that an increment in the size of the system will also multiply the number of users of the method). It turns out that QMC can be improved even further (a factor five in memory and CPU time) by an optimal non-unitary transformation that I have co-developed (in collaboration with Andrew Williamson)[27]. Our method is directly applicable to the study of realistic metallic systems a calculation that we plan to do for the first time.

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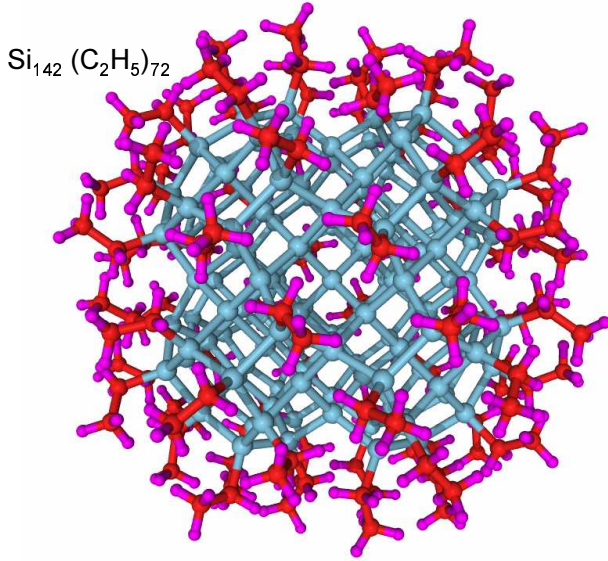


FIG. 7: Cluster of 142 Si atoms with a fully reconstructed surface terminated with ethyl groups. The organic passivation has an important influence on the optical gap and on the absorption spectra. Si and Ge quantum dots of this type are currently subject to an intense experimental research. (To appear in Ref. [27])

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